

CLAIMS

[1] A method for producing a compound single crystal comprising:
growing a compound single crystal by reacting a source gas with a
material solution,

5 wherein the single crystal is grown while stirring the material
solution to create a flow from a gas-liquid interface in contact with the source
gas toward an inside of the material solution.

[2] The method according to claim 1, using a single crystal production
apparatus comprising a heating unit and a closed pressure- and
10 heat-resistant container that is heated inside the heating unit,

wherein the source gas of the compound single crystal and other
materials are provided to the container and sealed in a pressurized
atmosphere,

the container is housed in the heating unit,

15 a material solution is prepared by heating the container in the
heating unit so that the other materials melt into a liquid, and

under these conditions, a single crystal is grown by reacting the
source gas with the material solution while the material solution is stirred.

[3] The method according to claim 2, wherein the single crystal is grown
20 by reacting the source gas with the material solution while the material
solution is stirred by rocking the container.

[4] The method according to claim 3, wherein the container is rocked by
rocking the heating unit.

[5] The method according to claim 2, wherein a crucible is set in the
25 container, and at least one of an inside and an inner wall of the crucible has
at least one selected from the group consisting of (A) impeller, (B) baffle, (C)
template, and (D) helical protrusions.

[6] The method according to claim 3, wherein the rocking is at least one
selected from the group consisting of a shift motion, a linear repetitive motion,
30 a pendular motion, a rotational motion, and a combined motion of any of

these motions.

[7] The method according to claim 2, wherein the other materials include a flux material.

[8] The method according to claim 3, wherein the single crystal production apparatus further comprises a source gas supply unit,
the source gas supply unit is connected to the container in which the other materials have been put, supplies the source gas to the container, and is separated from the container after supplying the source gas, and subsequently the container is rocked.

10 [9] The method according to claim 8, wherein the source gas supply unit is separated from the container after heating the container so that the other materials melt into a liquid and adjusting a pressure in the container.

[10] The method according to claim 2, wherein a pressure of the source gas in the container is reduced after formation of the single crystal.

15 [11] The method according to claim 2, wherein the single crystal production apparatus further comprises an auxiliary tank system for supplying the source gas, and the auxiliary tank system is connected to the container.

[12] The method according to claim 3, wherein the single crystal production apparatus further comprises a source gas supply unit, the source gas supply unit and the container are connected by a flexible pipe, and the container is rocked without being separated from the source gas supply unit.

[13] The method according to claim 1, wherein the source gas includes at least one of nitrogen and ammonia, other materials include at least one Group III element selected from the group consisting of gallium, aluminum, and indium and a flux material, and the single crystal formed in the material solution is a Group III nitride single crystal.

[14] The method according to claim 13, wherein the flux material includes at least one of an alkali metal and alkaline-earth metal.

30 [15] The method according to claim 13, wherein a template that comprises

a semiconductor layer expressed as a composition formula: $\text{Al}_u\text{Ga}_v\text{In}_{1-u-v}\text{N}$ ($0 \leq u \leq 1$, $0 \leq v \leq 1$, and $0 \leq u + v \leq 1$) is placed in a container beforehand.

[16] The method according to claim 15, wherein the material solution is prepared by heating in the container, the source gas is dissolved in the
5 material solution, and subsequently the template is immersed in the material solution.

[17] The method according to claim 15, wherein a crucible is set in the container, and the template is a sheet template and placed substantially upright on a bottom of the crucible.

10 [18] The method according to claim 17, wherein the container is rocked so that the material solution moves parallel to the sheet template.

[19] The method according to claim 7, wherein at least the flux material is removed from the container after the growth of the compound single crystal is finished.

15 [20] The method according to claim 19, wherein the material solution includes at least gallium and sodium, and a heating temperature of the material solution is 100°C (373 K) or more.

[21] The method according to claim 20, wherein the heating temperature is 300°C (573 K) or more.

20 [22] The method according to claim 20, wherein the heating temperature is 500°C (773 K) or more.

[23] The method according to claim 13, wherein a growth rate of the Group III nitride single crystal is 30 $\mu\text{m}/\text{hour}$ or more.

25 [24] The method according to claim 13, wherein a growth rate of the Group III nitride single crystal is 50 $\mu\text{m}/\text{hour}$ or more.

[25] The method according to claim 13, wherein a growth rate of the Group III nitride single crystal is 100 $\mu\text{m}/\text{hour}$ or more.

[26] The method according to claim 2, wherein a pressure of the source gas in the container is 5 atm ($5 \times 1.01325 \times 10^5 \text{ Pa}$) to 1000 atm (1000×1.01325

30 $\times 10^5 \text{ Pa}$).

[27] The method according to claim 2, wherein the heating unit is filled with an inert gas.

[28] The method according to claim 2, wherein when the other materials include gallium, the following formula (1) is satisfied:

$$5 \quad V \times (P/1.01325 \times 10^5) \times (T_1/T) > (X/2a) \times 22.4 \times 2 \quad (1)$$

where X (g) is a weight of gallium to be consumed, a (= 69.723) is an atomic weight of gallium, V (liter) is an internal volume of the container, P (Pa) is an ambient pressure during a growth process (formation of the single crystal), T (K) is a growth temperature, and T1 (K) is a temperature at which the other materials are weighed.

[29] The method according to claim 28, wherein the following formula (2) is satisfied instead of the formula (1):

$$V \times (P/1.01325 \times 10^5) \times (T_1/T) > (X/2a) \times 22.4 \times 5 \quad (2).$$

[30] The method according to claim 28, wherein the following formula (3) is satisfied instead of the formula (1):

$$15 \quad V \times (P/1.01325 \times 10^5) \times (T_1/T) > (X/2a) \times 22.4 \times 10 \quad (3).$$

[31] The method according to claim 2, wherein the single crystal production apparatus comprises a pipe for connecting the container in the heating unit and the outside of the heating unit, and the pipe has a structure that is likely to prevent aggregation of at least one of the material solution and the other materials.

[32] The method according to claim 31, wherein an inner diameter of the pipe is 3 mm or less.

[33] The method according to claim 31, wherein an inner diameter of the pipe is 2 mm or less.

[34] A single crystal production apparatus used for the method according to claim 2, comprising:

a closed pressure- and heat-resistant container;

a heating unit for housing the container; and

30 a rocking unit for rocking the container.

[35] The apparatus according to claim 34, wherein the container is rocked with the heating unit.

[36] The apparatus according to claim 34, wherein the rocking is at least one selected from the group consisting of a shift motion, a linear repetitive motion, a pendular motion, a rotational motion, and a combined motion of any of these motions.
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[37] The apparatus according to claim 34, wherein a crucible is set in the container, and at least one of an inside and an inner wall of the crucible has at least one selected from the group consisting of (A) impeller, (B) baffle, (C)
10 template, and (D) helical protrusions.

[38] The apparatus according to claim 34, wherein the container is housed in the heating unit to maintain a constant temperature.

[39] The apparatus according to claim 34, further comprising a source gas supply unit.

15 [40] The apparatus according to claim 39, wherein the container and the source gas supply unit are connected and separated freely.

[41] The apparatus according to claim 39, further comprising a flexible pipe,

20 wherein the container and the source gas supply unit are connected by the flexible pipe.

[42] The apparatus according to claim 34, further comprising an auxiliary tank system for supplying the source gas,

wherein the auxiliary tank system is connected to the container.